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Experimental electron Compton profile beyond the impulse approximation. Comparison of results on NH_3 and Ne (*)

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Résumé. — La partie symétrique du profil Compton du néon, déterminée par diffusion d'électrons de 25 keV, est comparée aux calculs théoriques suivant l'approximation de l'impulsion, et la partie antisymétrique est comparée aux prévisions qualitatives de Tavard pour des orbitales hydrogénoïdes. Les résultats antérieurs sur NH_3 sont rappelés et une tentative d'interprétation physique est donnée.

Abstract. — The symmetrical part of the neon Compton profile, obtained by 25 keV electron scattering is compared to impulse approximation calculations, and the antisymmetrical part is compared to a qualitative estimate by Tavard for hydrogenic orbitals. Previous results on NH_3 are recalled and a physical interpretation is attempted.

Within the framework of the impulse approximation (IA), the Compton profile (CP) of any target peaks at the energy-loss, E_{IA} , corresponding to the scattering by a free and stationary electron. The existence of a *Compton shift*, $\Delta E = E_{\text{exp}} - E_{\text{IA}}$, where E_{exp} is the observed energy-loss at the maximum of the CP, was established by Barlas *et al.* [1]. However, we have shown in recent experiments [2, 3] that this Compton shift is not sufficient to describe the deviations of the NH_3 CP from the IA predictions. The purpose of this note is to report similar deviations as recently measured for neon and to compare them to the NH_3 results which will be briefly recalled.

The CP of NH_3 , $J(q, K)$, was determined at different scattering angles ⁽¹⁾, and it was found to depend not only on the energy-loss E , through

$$q = (2E - K^2)/2K \quad (2),$$

but also on the momentum transfer K , while the corresponding quantity $J_{\text{IA}}(q)$, as predicted according to the IA, is only a function of q . However there are two K -ranges where the experimental profile is practically independent of K ; for $K > 6$ a.u., one gets a *total profile* $J^{\text{T}}(q)$ due to electrons ejected from all orbitals of the molecule; for $3 \text{ a.u.} < K < 5 \text{ a.u.}$, it is a *valence profile* $J^{\text{V}}(q)$ due to electrons ejected in collisions where the $1a_1$ electrons of NH_3 are excluded. In each of these ranges, the symmetrical part $J_{\text{s}}(q) = (J(q) + J(-q))/2$ is in good agreement with the theoretical corresponding IA profile, respectively $J_{\text{IA}}^{\text{T}}(q)$ and $J_{\text{IA}}^{\text{V}}(q)$; the antisymmetrical part $J_{\text{a}}(q) = (J(q) - J(-q))/2$ is then a good representation of the second order term of the profile, that is total profile for $K > 6$ a.u. and valence profile for $3 \text{ a.u.} < K < 5 \text{ a.u.}$; by subtraction, one can obtain the symmetrical and the antisymmetrical parts, $J_{\text{s}}^{1a_1}(q)$ and $J_{\text{a}}^{1a_1}(q)$, corresponding to ionizing collisions on the NH_3 $1a_1$ orbitals formed by the nitrogen $1s$ orbitals.

Electron CP's have been obtained for neon from energy-loss spectra of 25 keV electrons scattered in the angular range corresponding to momentum transfer values from 1 to 15 atomic units [4]. Figure 1 shows, for two scattering angles, the cross section doubly differential in angle and energy-loss, $d^2\sigma/dE d\Omega$. For $K < 7$ a.u. (Fig. 1a), the main profile, up to $E \approx 870$ eV, is due to the scattering

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⁽¹⁾ Bonham has shown [8] that, along a profile at constant scattering angle, the momentum transfer K is very close to its value at the maximum of the profile.

⁽²⁾ All quantities are here expressed in atomic units. The expression for q in Ref. [2] is correct if the energy is evaluated in Rydberg (1 a.u. = 2 Ry).

Fig. 1. — Cross sections, $d^2\sigma/d\Omega dE$, differential with respect to the angle and the energy-loss for inelastic scattering of electrons from : a) the L-electrons, and b) all the electrons of neon.

by the L-electrons (2s and 2p) of the neon atom; it is a valence profile. For $K > 10$ a.u. (Fig. 1b), all neon electrons (1s, 2s, 2p) are involved in the scattering process; it is a total profile. A profile is usually represented by $J(q, K) = \frac{K^5}{2} \frac{k_0}{k} \frac{d^2\sigma}{dE d\Omega} (k_0$ and k are the initial and final momenta of the colliding electron). Figure 2 represents the maximum value of each profile, $J_{\max}(q, K)$, as a function of K . In the framework of the IA, this maximum is simply the expectation value $N \langle p^{-1} \rangle$ relative to the momenta p of the N target electrons contributing to the profile. There are two plateaus in the K -ranges 4 a.u. $< K < 8$ a.u. and 8 a.u. $< K$, where the IA is a good first order approximation, respectively for the valence profile ($N = 8$, first range) and the total profile ($N = 10$, second range) [3].

Fig. 2. — The maximum value, $J_{\max}(q, K)$, of the Ne electron CP versus momentum transfer K .

Using the mean profile, $J(q)$, as determined in each of these two regions for K values not too close to the limits, we have calculated $J_s(q)$ and $J_a(q)$. $J_s(q)$ is in good agreement with $J_{IA}(q)$ calculated in the framework of the IA [5], with deviations smaller than respectively 2% for the valence profile up to $q = 1.5$ a.u. and 1.9% for the total profile up to $q = 2.0$ a.u.. The antisymmetrical part $J_a(q)$ is represented in figure 3 for the total profile, the valence profile, and their difference which gives the K-shell profile (1s orbitals). These curves are in accordance with the theoretical qualitative predictions of Tavard for hydrogenic orbitals [6].

Fig. 3. — Antisymmetrical parts of the total, valence, and 1s Ne electron CP's, respectively $J_a^T(q)$, $J_a^V(q)$ and $J_a^1s(q)$. The incident electron energy is 25 keV.

The NH_3 antisymmetrical experimental profiles [3] (Fig. 4) behave differently, especially the nitrogen K-shell profile, whose sign is opposite to that of neon,

Fig. 4. — Same as in figure 3 but for NH_3 , with an incident electron energy of 35 keV.

and whose magnitude is about fifty times lower at the maximum; the antisymmetrical total and valence profiles are then very close to each other.

According to Tavard's interpretation, the electron CP, $J(q, K)$, can be written [7] as the sum of a first term which exactly represents the IACP, $J_{IA}(q)$, and of a second order term related to some integrals of the potential interaction type (see Ref. [7] for the exact equations). Qualitative predictions can be obtained by assuming that the energy-losses are large compared to the ionization energies involved, and mainly by taking into account the action of localized charges on the ejected electrons, i.e. by neglecting in a first step the electron-electron interactions compared to the electron-nucleus interactions. The electron-electron interactions are of opposite sign compared to the electron-nucleus ones and they are not localized, hence they are equivalent to a lowering of the nucleus effective charge. For neon, this lowering does not exceed the number of non-ejected electrons, $Z - 1$, the effective charge is then positive and larger than 1. For NH₃, the lowering of the effective charge is due to the $Z - 1$ electrons of N and to the transfer of electronic charge from the three hydrogen to the central atom occurring during the chemical bond formation;

the six bond electrons (3p electrons from N and 3 electrons from H) being quite localized may then sufficiently lower or even make slightly negative the nitrogen effective charge. This can explain the difference in sign and order of magnitude between the NH₃ and the Ne antisymmetrical profiles. It is also to be noted that a K-electron of N contributing to the $1a_1$ -NH₃ profile is at a large distance from the H nuclei ($r_{NH} \approx 1 \text{ \AA}$; radius of the K-orbit $r_{1sN} \approx 0.1 \text{ \AA}$), and hence the action of these nuclei are negligible.

It is to be noted that in this research, profiles could be obtained only in experimental conditions where the energy-loss at the maximum of the profile is not very large compared to the ionization energy of the concerned shell (10 times for the valence shell, but only 3 times for the K-shell); this is not very favourable for theoretical interpretation.

Experimental work on H₂O and CH₄ as well as further theoretical studies seem to be desirable in this domain.

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